

UNMARKED VERSION OF AMENDMENTS

1. **(AMENDED)** A process for the preparation of low molecular weight linear alpha olefins having 4 to 24 carbon atoms, comprising ~~oligomerising~~ oligomerizing ethylene in an inert aliphatic or aromatic solvent in the presence of a catalyst including at least two components, a first component selected from the group consisting of zirconium alkoxide and zirconium aryloxide in association with free alcohol in a ratio of about 1:0.33 to about 1:2.3, and a second component selected from the group consisting of an alkyl aluminum and/or alkyl aluminum halide ~~component~~.
2. **(ORIGINAL)** A process as claimed in claim 1 wherein the process is carried-, out under a continuous supply of ethylene and under agitation.
3. **(ORIGINAL)** A process as claimed in claims 1 wherein the process is performed in semi-continuous mode with ethylene being fed continuously during each period of the process.
4. **(ORIGINAL)** A process as claimed in claim 1 wherein the catalyst system comprises of at least two components, the first component comprising of zirconium (IV) alkoxide or carboxylate and the second component comprising of triethylaluminum and/or ethylaluminum sesquichloride.
5. **(ORIGINAL)** A process as claimed in claim 1 wherein the catalyst is of the formula $Zr(OR)_4-Et_3Al$ wherein R is alkyl or aryl.
6. **(ORIGINAL)** A process as claimed in claim 1 wherein the catalyst is of the formula $Zr(OR)_4-Et_3Al_2Cl_3$ wherein R is alkyl or aryl.
7. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 1 wherein the catalyst is of the formula $Zr(OR)_4Et_3Al$ and/or $Zr(OR)_4Et_3Al_2Cl_3$ wherein R is alkyl or aryl.
8. **(ORIGINAL)** A process as claimed in claim 5 wherein Et_3Al is reacted with $Zr(OR)_4$ in the mole ratio of 10:1 to 60:1.

9. **(ORIGINAL)** A process as claimed in claim 6 wherein $\text{Et}_3\text{Al}_2\text{Cl}_3$ is reacted with $\text{Zr}(\text{OR})_4$ in the mole ratio of 10:1 to 60:1.
10. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 7 wherein Et_3Al and/or $\text{Et}_3\text{Al}_2\text{Cl}_3$ is reacted with $\text{Zr}(\text{OR})_4$ in the mole ratio of 10:1 to 60:1.
11. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 4 wherein zirconium alkoxide is in association with the free alcohol in a concentration ratio range of 1:0.33 to 1:1.3.
12. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 7 wherein when both Et_3Al and $\text{Et}_3\text{Al}_2\text{Cl}_3$ are used, the Et_3Al diluted in solvent is initially charged into the reactor and then $\text{Et}_3\text{Al}_2\text{Cl}_3$ and other catalyst components are added therein.
13. **(ORIGINAL)** A process as claimed in claim 1 wherein the ethylene pressure is in the range of 18 to 38 kg/cm^2 .
14. **(AMENDED)** A process as claimed in claim 1 wherein the ~~oligomerisation~~ oligomerization is carried out at a temperature in the range of 80°C to 140°C.
15. **(ORIGINAL)** A process as claimed in claim 1 wherein, the process is carried out for a time period in the range of 1 hour to 3 hours.
16. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 1 wherein, the solvent used is selected from the group consisting of: cyclohexane, toluene and n-octane.
17. **(ORIGINAL)** A process as claimed in claim 2 wherein the reaction is carried out at an agitator speed of 300 to 1000 rpm.
18. **(ORIGINAL)** A process as claimed in claim 1 wherein, the zirconium component is selected from the group consisting of zirconium tetra cresylate, zirconium tetra dimethyl phenolate, zirconium tetra n-butoxide, zirconium tetra iso-propoxide, zirconium tetra n-propoxide, zirconium tetra butyrate and zirconium tetra isobutyrate.

19. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 1 wherein said catalyst includes a thiophene as a third component to reduce chain growth.
20. **(AMENDED)** A process for the preparation of low molecular weight linear alpha olefins having 4 to 24 carbon atoms, comprising ~~oligomerising~~ oligomerizing ethylene in an inert aliphatic or aromatic solvent in the presence of a catalyst $\text{Zr(OR)}_4\text{-Et}_3\text{Al}$ and/or $\text{Zr(OR)}_4\text{-Et}_3\text{Al}_2\text{Cl}_3$ wherein R is alkyl or aryl, at a pressure is in the range of 18 to 38 kg/cm^2 , a temperature in the range of 80°C to 140°C for from 1 hour to 3 hours.
21. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 17, wherein the mole ratio of Et_3Al and/or $\text{Et}_3\text{Al}_2\text{Cl}_3$ to Zr(OR)_4 is 10:1 to 60:1.
22. **(ORIGINAL)** A process as claimed in claim 17 wherein the reaction is carried out at an agitator speed of 300 to 1000 rpm.
23. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 17 wherein said catalyst includes a thiophene as a third component to reduce chain growth.
24. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 17 wherein said solvent is selected from the group consisting of: toluene, n-Octane and cyclohexane.
25. **(AMENDED)** A process for the preparation of low molecular weight linear alpha olefins having 4 to 24 carbon atoms, comprising ~~oligomerising~~ oligomerizing ethylene in an inert aliphatic or aromatic solvent in the presence of a catalyst $\text{Zr(OR)}_4\text{-Et}_3\text{Al}_2\text{Cl}_3$ wherein R is alkyl or aryl, at a pressure is in the range of 18 to 38 kg/cm^2 , a temperature in the range of 80°C to 140°C for from 1 hour to 3 hours.
26. **(ORIGINAL)** A process as claimed in claim 25, wherein the mole ratio of $\text{Et}_3\text{Al}_2\text{Cl}_3$ to Zr(OR)_4 is 10:1 to 60:1.
27. **(ORIGINAL)** A process as claimed in claim 25 wherein the reaction is carried out at an agitator speed of 3 00 to 1000 rpm.
28. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 25 wherein said catalyst includes a thiophene as a third component to reduce chain growth.

29. **(PREVIOUSLY PRESENTED)** A process as claimed in claim 25 wherein said solvent is selected from the group consisting of: toluene, n-Octane and cyclohexane.
30. **(CANCELED)**

III. REMARKS

RESPONSE TO REJECTIONS

In the Claims:

- **Claim Rejections under 35 U.S.C. § 112, First Paragraph**

- **The Examiner's Position:**

The Examiner has rejected claim 30 under 35 U.S.C. § 112, first paragraph, for "containing subject matter which was not described in the specification in such a way as to reasonably convey to one skilled in the relevant art that the inventor(s), at the time the application was filed, had possession of the claimed invention" (paragraph 2, page 2, of the Office Action). The Examiner asserts that claim 30 has a negative limitation which "must have positive support from the specification." The Examiner finds no support for such negative limitation in the specification.

- **Applicants' Position:**

Applicants respectfully traverse the Examiner's position on the basis that a person of ordinary skill in the art would at the time the application was filed understand from the disclosure that the applicants had possession of the invention made in claim 30. However, as claim 30 is cancelled herein without prejudice, the Applicants assert that such rejection is mooted.

- **Claim Rejections under 35 U.S.C. § 112, Second Paragraph**

- **The Examiner's Position:**

The Examiner has rejected claims 12 and 30 under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which the applicant regards as the invention (paragraph 4, page 2, of the Office Action). With respect

to claim 30, the Examiner asserts that it is unclear whether zirconium halide is excluded from the process or not (paragraph 5, page 2, of the Office Action). The Examiner asserts that with respect to claim 12 it is unclear whether the claimed process requires using both Et_3Al and $\text{Et}_3\text{Al}_2\text{Cl}_3$.

- **Applicants' Position:**

Applicants respectfully traverse the Examiner's position on the basis that a person of ordinary skill in the art would understand the claims as drafted as particularly pointing out and distinctly claiming the subject matter thereof. However, cancellation of claim 30 without prejudice, herein, moots the Examiner's rejection. In regard to claim 12, Applicants assert that claim is clearly directed to an embodiment of the subject matter asserted in claim 1 wherein "both Et_3Al and $\text{Et}_3\text{Al}_2\text{Cl}_3$ are used" (emphasis added). Claim 12 depends from claim 1 which recites a process wherein the second component from the group consists of an alkyl aluminum and/or alkyl aluminum halide component" (emphasis added). Applicants therefore respectfully request that the Examiner withdraw his 35 U.S.C. §112, second paragraph, rejections.

- **Claim Rejections under 35 U.S.C. § 103(a):**

- **The Examiner's Position:**

The Examiner has rejected claims 1 - 3, 13 - 17, 18 and 30 under 35 U.S.C. §103(a) as being unpatentable over U.S. Patent No. 4,409,414 to Langer Jr. (the "Langer Jr. '414 reference") (paragraph 4, page 3, of the Office Action), claim 19 as being unpatentable over the Langer Jr. '414 reference in view of U.S. Patent No. 5,260,500 to Shiraki *et al.* ("Shiraki *et al.* '500 reference"), and claims 4 - 12, 20 - 22, 24 - 27 and 29 in light of the Langer Jr. '414 reference in view of U.S. Patent No. 4,855,525 to Young *et al.* ("Young *et al.* '525 reference"), and claims 23 and 28 in light of the Langer Jr. '414 reference in view of U.S. Patent No. 4,855,525 to Young *et al.* ("Young *et al.* '525 reference") and further in view of the Shiraki *et al.* '500 reference.

The Examiner asserts that the Langer Jr. '414 reference "discloses a batch/continuous process of making alpha-linear olefins having applicants' claimed range of number of carbons by oligomerizing ethylene in the presence of a catalyst containing zirconium alkoxide such as

Zr(OBu)₄ and alkyl aluminum halide, in the presence of a diluent such as toluene under the condition of a temperature ranging from below 125⁰C, a pressure ranging from above 50 psia such as 500 psia, during the applicants' claimed time and high-speed stirring" (paragraph 5, page 3 – paragraph 1, page 4, of the Office Action). The Examiner also asserts that Langer Jr. '414 reference discloses that the process is operated in the presence of alcohol to enhance the polymerization process, and that the reference teaches that the amount of alcohol employed effects the molecular weight of the product (paragraph 3, page 4, of the Office Action). In regard to claims 1 – 3, 13 – 17, 18 and 30, the Examiner asserts that the selection of the appropriate amount of added alcohol to obtain product having the molecular weights recited was nothing more than routine experimentation (paragraph 4, page 4, of the Office Action). In regard to those claims asserting stirring speed, the Examiner likewise asserts that such stirring speeds would be selected by one of ordinary skill in the art at the time the invention was made (paragraph 3, page 5, of the Office Action).

Arguing that the Shiraki *et al.* '500 reference discloses that "in a process for producing a linear alpha olefins that "it is effective to add to the catalyst a sulfur compound such as thiophene to improve the purity of the linear alpha olefins" (paragraph 5, page 5, of the Office Action), the Examiner asserts that with respect to claim 19's recitation of thiophene that "[i]t would have been obvious to one having ordinary skill in the art to modify the Langer process by adding an amount of thiophene to increase the purity of the product" (paragraph 7, page 5, of the Office Action). Likewise, the Examiner argues that the thiophene recitation of claim 28, which depends on claim 17, and ultimately claim 2, to be obvious in light of the Shiraki *et al.* '500 reference for the same reason (paragraphs 3 – 5, page 6, of the Office Action).

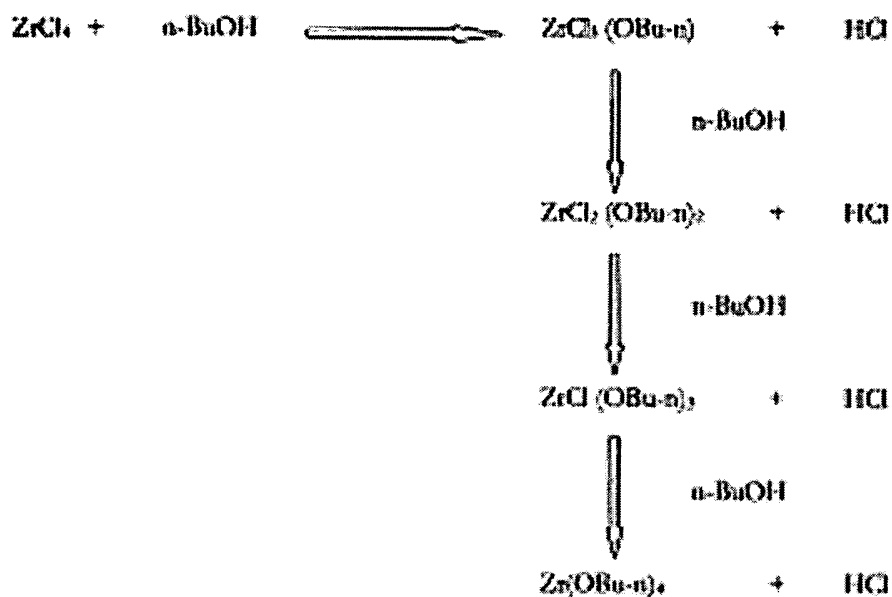
Recognizing that claims 4 – 12, 20, 21, 25 and 26 disclose the use of the aluminum compounds that are not disclosed in the Langer Jr. '414 reference, the Examiner asserts that the Young *et al.* '525 reference discloses "equivalent" aluminum compounds. The Examiner rejects arguments said to made in Applicants' "Amendment and Response to Office Action of June 19, 2002" (paper no. 6) to the effect that the compounds disclosed in the Young *et al.* '525 reference are aluminum compounds having an adduct of zirconium tetrahalide as non-persuasive given the Examiner's assertion that the aluminum compounds in the Young *et al.* '525 reference are

equivalent to those recited in the claims (paragraph 5, page 7, of the Office Action). The Examiner states that it would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer Jr. '414 process by using $R_3Al_2X_3$ and AlR_3 as the aluminum component for the Langer catalyst "since it is expected that using any equivalent aluminum compounds disclosed by Young would yield similar results" (paragraph 2, page 6, of the Office Action). With respect to claim 28 which depends from claim 25, the Examiner again finds the missing thiophene recitation of the claim to be obvious in light of the Shiraki *et al.* '500 reference.

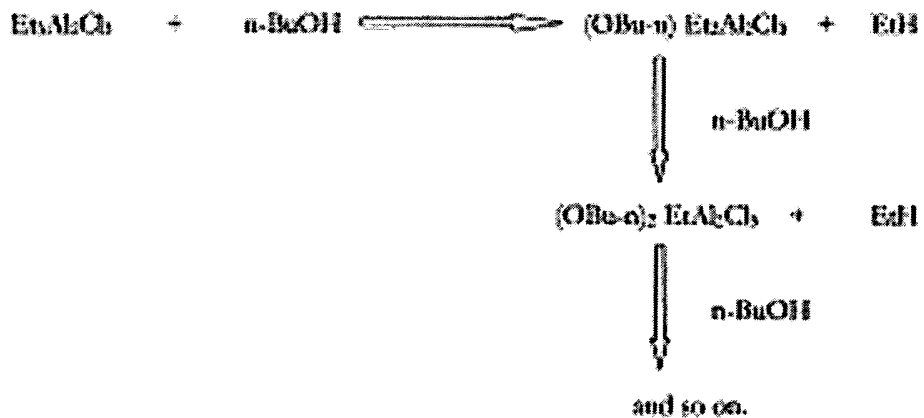
Applicants' Position:

Applicants respectfully traverse the Examiner's 35 U.S.C. § 103(a) rejections based in part on the failure of the Examiner to recite adequate motivation for combining the references in the manner indicated, and based in part on the inappropriate use by the Examiner of "hindsight reasoning" in an attempt to "approximate" the present invention.

First, Applicant respectfully asserts that the Examiner has mis-read the disclosures of the Langer Jr. '414 reference. The '414 patent discloses an *in situ* reaction of a transition metal halide with an alcohol. Under conditions described in the patent a hydrogen halide is produced. For example, in Example 7, $ZrCl_4$ is reacted with n-butanol *in situ*. This results in the formation of a mixture of $ZrCl_4$, $ZrCl_3(OBu-n)$, $ZrCl_2(OBu-n)_2$, $ZrCl(OBu-n)_3$, $Zr(OBu-n)_4$ **and** HCl , with the conversion of $ZrCl_4$ to $Zr(OBu-n)_4$ never going to completion in the absence of a suitable base.



Further, as unreacted free BuOH is always present in the solution, it reacts with the aluminum alkyl halide compound of the Langer Jr. '414 reference form a number of different species. As would be understood by one of ordinary skill in the art, because of the varied species produced, reaction with the transition metal complex system is entirely unpredictable.



The halide ligand required in the Langer Jr. '414 reference (See, col. 4, lines 63 – 68; col. 5, lines 1 – 68) is completely absent in the zirconium metal complex used in the instant invention. Respectfully the Examiner takes several leaps which simply are not supported in the art in a hindsight attempt to equate compositions recited in the present invention with those disclosed in the Langer Jr. '414 reference. As would be understood by one of ordinary skill in the art, there is no assumption of predictability in catalyst systems such as those disclosed in the Langer Jr. '414 reference and claimed in the instant invention. The Langer Jr. '414 reference simply does not provide any teaching or suggestion that a halide ligand-containing zirconium complex which is generated *in situ* is equivalent to a non-halide containing zirconium complex which is not generated *in situ*. Nor would one of ordinary skill in the art *a priori* have any basis for equating the same.

The very unpredictability of such catalyst systems is illustrated in Run 1 of Table IV of the Langer Jr. '414 reference. As seen, $\text{AlEt}_2\text{Cl} + \text{Zr}(\text{OPr})_4$ was inactive, while $\text{AlEt}_2\text{Cl} + \text{AlEtCl}_2 + \text{Zr}(\text{OPr})_2$ in a mole ratio of 0.2:0.6:0.1 was evidenced to be active. $\text{Zr}(\text{OBu})_4 \cdot \text{BuOH}$ exists as a trimer similar to $\text{Zr}(\text{OPr})_4$ (Ref: Bradley *et al.*, *Metal alkoxide*, Academic Press, p. 63, 67, 1978). However the presence of toluene and similar solvents, the complex is in the form of a dimer (Ref: Vaartstra, *et al.*, *Alcohol adducts of a/k oxides: Intramolecular hydrogen bonding as a general structural feature*, Inorg. Chem, 1990, 29, 3126 - 3131). This dimer molecule is considered by the skilled artisan as different from $\text{Zr}(\text{OBu})_4$. It is also understood in the art that alcohol free $[\text{Zr}(\text{OPr})_4]_m$ and $\text{Zr}(\text{OPr})_4 \cdot \text{PrOH}$ have different properties. It therefore follows that *in situ* generated $\text{ZrCl}_4 + n\text{-BuOH}$ is different from the defined $\text{Zr}(\text{OBu})_4 \cdot \text{XbuOH}$ (where $X = 0.3, 0.4, 1$)(a halide-free zirconium metal complex as used in the present invention) are different. In short, there simply is no predictability in the catalyst system.

The catalyst system of the Langer Jr. '414 reference does not employ either AlEt_2Cl and/or AlEtCl_2 as used in the Langer Jr. '414 reference. Rather compounds such as Et_3Al and/or $\text{Et}_3\text{Al}_2\text{Cl}$ are employed. Thus in their very constituents, the two catalyst systems are different.

As stated above there is simply no basis for making the leap that $\text{ZrCl}_4 + n\text{-BuOH}$ and

$\text{Zr}(\text{OBu})_4$.XbuOH are equivalents in terms of their catalytic activity. Nor is there any basis for assuming that it is immaterial in respect of catalytic activity that the zirconium complex of the Langer Jr. '414 reference is generated *in situ* and that the system that exists in the reactor described in the Langer Jr. '414 reference is immaterial. Further, there is no basis for asserting that one of ordinary skill in the art would ignore the teachings of the Langer Jr. '414 reference in regard to the focus on killing of catalyst activity.

In regard to claims 4 – 12, 20 – 22, 24 – 27, and 29 that have been rejected as being obvious in light of the Langer Jr. '414 reference in view of the Young *et al.* '525 reference, respectfully Applicants once more assert that the Examiner has erred in simplistically assuming equivalence where one of ordinary skill in the art would not.

Applicants reiterate the differences between the teachings of the Langer Jr. '414 reference and the present invention which are set forth *supra*. Likewise in regard to the Young *et al.* '525 reference the compounds taught therein also are known to have significantly different properties than those used in the present invention. For example, the Young *et al.* '525 reference teaches adducts of ZrCl_4 with ketones, esters, nitriles, etc. This is a different class of compounds altogether from $\text{Zr}(\text{OR})_4$ (e.g., R = butyl, phenyl). For example, ZrCl_4 reacts with alcohols to provide a partially exchanged alkoxide. $\text{Zr}(\text{OBu})_4$, however, does not provide such alkoxides. The catalysts of the Young *et al.* '525 reference and those of the instant invention belong to different class of compounds and give a different reactivity. Further, as indicated in the "Amendment and Response to Office Action of June 19, 2002" the Young *et al.* '525 reference discloses aluminum compounds being strictly used with respect to a catalyst having adduct of zirconium tetrahalide as a component, certainly not referenced in the claims. In short, there is simply no motivation to combine the teachings of the Young *et al.* '525 reference with those of the Langer Jr. '414 reference.

The Examiner recites to the Shiraki *et al.* '500 reference for the teaching of the addition of thiophene to the catalyst in rejecting claims 19, 23 and 28. The Applicants respectfully disagree with the Examiner's reasoning with respect to this reference as well.

It is important to note that the Shiraki *et al.* '500 reference differs from the instantly

claimed embodiments of the invention in that the catalyst system is also based on the $ZrCl_4$ system and the catalyst system, as the catalyst system in the Langer Jr. '414 reference, is generated *in situ*. In the Shiraki *et al.* '500 reference, $ZrCl_4$ and alkylaluminums are reacted first to provide a bridging complex. When butanol is added to this, it attaches Zr-Cl and Al- C_2H_5 bonds simultaneously. Reactivity of alkylaluminum with alcohol leads to the formation of aluminum alkoxides (See, e.g., Kunicki *et al.*, *Studies on the reaction of triethylaluminum with methyl alcohol*, Bull. Polish Acad. Sci. Chem., 33 No. 5 — 6, 1985). Thus, one of ordinary skill in the art is faced with a complex system, where the identity of the active species for the oligomerization reaction is difficult to predict/fathom.

While the Shiraki *et al.* '500 reference acknowledges that it was known at the time of the reference that sulfur compounds such as thiophene could be added to a catalyst system to improve the purity of the linear α -olefin production, it specifically teaches away from the use of thiophene in the system since the focus in this reference is on replacing the sulfur compound by an alcohol. Such is suggested to avoid the potential contamination of the linear α -olefin by the sulfur compound (column 1 lines 29 — 35 of the Shiraki *et al.* '500 reference).

Given the disclosures of the Shiraki *et al.* '500 reference, the Langer Jr. '414 reference, and the Young *et al.* '525 references, as discussed above, there is simply no motivation for combining them all together, or one reference with another reference. Therefore, it is respectfully requested that the Examiner withdraw the outstanding rejections with respect to at least claims 19, 23 and 28 which rest upon such a combination. The lack of motivation for combination is particularly so, since the catalyst systems in the Shiraki *et al.* '500 reference and the Langer Jr. '414 reference are not predictable *per se*. Further contrary to the Examiner's assertions, the Shiraki *et al.* '500 reference actually teaches away from the use of thiophene as an additive — replacing it with alcohol. Further the Langer Jr. '414 reference focus on the use of a base to kill catalyst activity in a designated time post-completion of the oligomerization reaction also teaches away from the present invention.

In light of such arguments with respect to the non-obviousness of the embodiments of the invention presently asserted, it is respectfully asserted that such 35 U.S.C. §103 rejections should be withdrawn with respect to claims 1 — 3, 4 — 12, 13 — 17, 18, 19, 20 — 22, 24 — 27, 29 and 30.

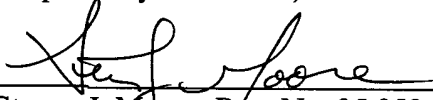
Applicants respectfully assert that all pending claims in this application are in condition of allowance and are not taught, suggested, or made obvious by the prior art of record. Applicants respectfully request an early notice of allowance with respect to all claims that are presently pending.

CONCLUSIONS

In view of the above, it is respectfully submitted that the subject matter of the pending claims which were rejected under 35 U.S.C. §§ 103(a), and 112, first and second paragraph, are patentable over the references of record. An early notice of allowance is therefore earnestly requested.

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Respectfully Submitted,


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